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Modification of spectral characteristics and optogalvanic response in neon hollow cathode under laser excitation

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Abstract. The changes in emission characteristics of a neon hollow cathode discharge by resonant laser excitation of $1s_5 \rightarrow 2p_2$ and $1s_5 \rightarrow 2p_4$ transition have been studied by simultaneously monitoring the optogalvanic effect and the laser induced fluorescence. It has been observed that resonant excitation causes substantial variation in the relative intensities of lines in the emission spectrum of neon discharge.

Keywords. Optogalvanic spectroscopy; hollow cathode discharge.

PACS Nos **82·80; 52·25**

1. Introduction

The selective perturbation of the energy level population using lasers of high spectral brightness, produces some noticeable modifications in the discharge characteristics. Laser optogalvanic (OG) effect [1], which is the change in impedance of a discharge, and laser induced fluorescence are the consequences of this perturbation. Investigations on simultaneous monitoring of OG effect and fluorescence phenomena in a neon discharge have been reported by a few workers [2–4]. Such studies give information regarding population of various levels, electron density in the discharge and the details of ionization processes that generate the OG effects. In this work, we present the changes in emission characteristics which occur simultaneously with the OG effect as a result of laser excitation of $1s_5 \rightarrow 2p_2$ and $1s_5 \rightarrow 2p_4$ transition in a neon hollow cathode discharge (hcd).

2. Experimental

A frequency stabilized ring dye laser (Spectra Physic-380D) pumped by argon ion laser (Spectra Physic-171) was used as the excitation source. Wavelength of the laser was exactly tuned on the line centre of the neon transition ($1s_5 \rightarrow 2p_2$, $1s_5 \rightarrow 2p_4$) where the OG signal amplitude is a maximum. The wavelength was measured with an accuracy of $0{\cdot}01\,\mathrm{cm}^{-1}$ using a wavemeter (Burliegh WA-20 VI). A commercial Ne/Mo hollow cathode lamp with a gas pressure of 13·3 mBar (Cathodean UK) was used as the discharge source and the laser beam was focused into it. A 0·5 m monochromator (Jarrell–Ash) was used for recording emission spectrum with and without laser excitation up to a discharge current of 5 mA. The PMT output was processed by a lock-in amplifier and a computer was used for the data acquisition. Since the

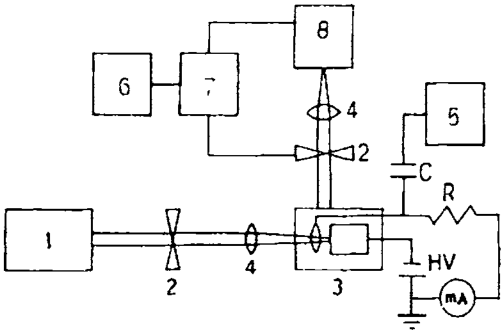

Figure 1. Experimental set-up (1. single mode ring dye laser, 2. chopper, 3. Ne–Mo hollow cathode, 4. lens, 5. CRO, 6. computer, 7. lock-in amplifier, 8. monochromator, $R = 45\,K\Omega$, $C = 0{\cdot}1\,\mu F$).

range of current used for the operation of the discharge was very low, Mo atomic lines were not observed in the emission spectrum. The neon transitions within the scanning range of the monochromator were identified in all cases. The OG signal which corresponds to an increase in the discharge impedance during laser excitation, was measured using an oscilloscope. A schematic of the experimental set up is shown in figure 1.

3. Results and discussion

a) *Energy level diagram of neon*

The first excited state of neon ($2p^5 3s$) has four components designated as $1s_i (i = 2$, 3, 4 and 5) two of which ($1s_3$ and $1s_5$) are metastables with a long life time. The next excited state configuration is $2p^5 3p$ giving rise to ten states represented as $2p_j$ ($j = 1$ to 10) which are coupled radiatively to the $1s_i$ states [5].

b) *Laser-induced fluorescence spectrum*

Laser excitation of $1s_5 \rightarrow 2p_2$ *transition (588·2 nm)*

The emission and the laser induced fluorescence spectrum of the neon hollow cathode at a discharge current of 5 mA by laser excitation of $1s_5 \rightarrow 2p_2$ transition in neon are shown in figure 2. This shows that laser absorption produces a noticeable change in the emission characteristics of the discharge (table 1). These changes are mainly due to non-radiative decay from $2p_2$ state to $2p_j$ ($j \neq 2$) state and also due to the coupling of these levels to the ground state. Measurements of variation in emission intensity of $2p_2 \rightarrow 1s_i$ ($i = 2, 3, 4$ and 5) transitions as a function of the discharge current (figure 3) show that in the presence of laser beam, the emission intensities are non-linear functions of current while in the absence of laser excitation they have a linear dependence. The enhancement in the intensity of these transitions during laser excitation indicates that the population of the $2p_2$ state increases considerably. The emission intensity of $2p_1 \rightarrow 1s_2$ also shows slight increase under laser irradiation due to collisional excitation of atoms from $2p_2$ to $2p_1$ level. It has been established that such collisional excitation or de-excitation cross-section between two levels with energy difference ΔE varies as $\exp(-\Delta E/kT)$. This implies that atoms in $2p_2$ level

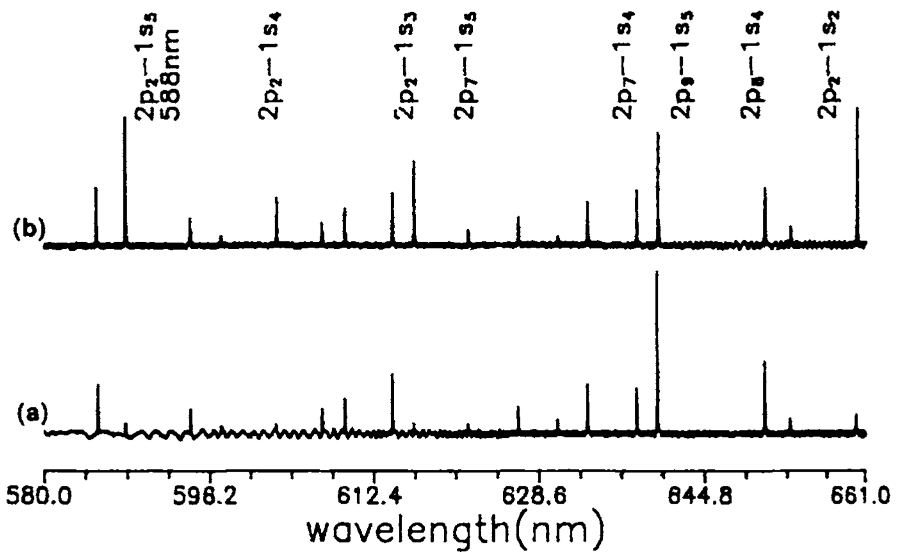

Figure 2. a) Emission spectrum of neon discharge at 5 mA and b) the same under laser irradiation at 588·2 nm.

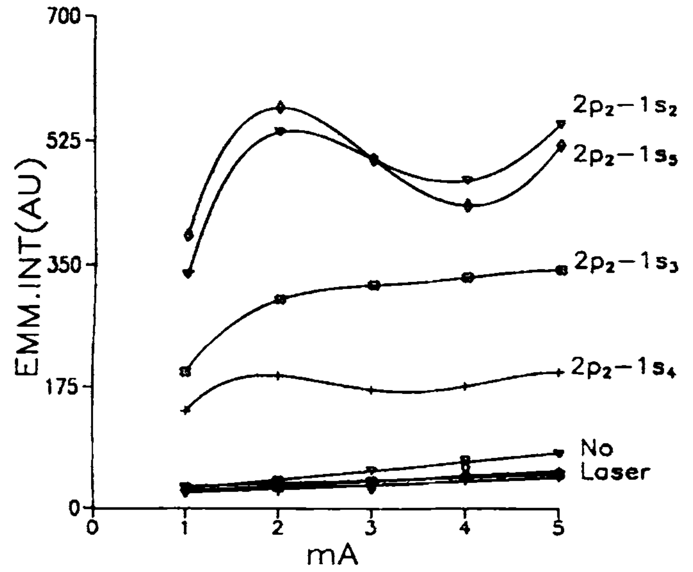

Figure 3. Dependence of the emission intensity for $2p_2 \rightarrow 1s_i$ ($i = 2, 3, 4, 5$) transitions on discharge current.

can de-excite to nearby sub levels ($j = 3, 4, 5$) while such collisional de-excitations to $2p_j$ levels with $j = 6, 7, 8, 9, 10$ will be less probable. It should be noted that population of $1s_5$ level will decrease due to absorption of laser light and hence the $2p_j$ manifolds arising through electron collision from $1s_5$ level viz,

$$Ne(1s_5) + e^- \rightarrow Ne(2p_j) + e^-$$

Table 1. Change in emission intensities under laser excitation at 588·2 nm and 594·5 nm with a discharge current of 5 mA.

Wavelength (nm)	Transition	$(I_L - I)/I$[a]	
		$1s_5 \to 2p_2$[b]	$1s_5 \to 2p_4$[c]
585·2	$2p_1 \to 1s_2$	0·17	~0
588·2	$2p_2 \to 1s_5$	9·23[d]	0·23
594·5	$2p_4 \to 1s_5$	0·09	22·0[d]
597·5	$2p_5 \to 1s_5$	0·12	0·17
603·0	$2p_2 \to 1s_4$	3·33	~0
607·4	$2p_3 \to 1s_4$	−0·03	1·50
609·6	$2p_4 \to 1s_4$	0·03	5·06
614·3	$2p_6 \to 1s_5$	−0·12	0·07
616·3	$2p_2 \to 1s_3$	5·79	~0
621·7	$2p_7 \to 1s_5$	0·55	0·02
626·6	$2p_5 \to 1s_3$	0·04	1·09
630·4	$2p_6 \to 1s_4$	−0·30	−0·16
633·4	$2p_8 \to 1s_5$	−0·10	0·06
638·3	$2p_7 \to 1s_4$	0·18	0·21
640·2	$2p_9 \to 1s_5$	−0·29	−0·53
650·6	$2p_8 \to 1s_4$	−0·18	−0·39
653·2	$2p_7 \to 1s_3$	0·14	−0·04
659·9	$2p_2 \to 1s_2$	5·85	~0
667·8	$2p_4 \to 1s_2$		6·12

[a] I_L and I are emission intensities with and without laser irradiation.

[b] 10 μm and [c] 5 μm—slit width of the monochromator used for recording the spectrum.

[d] Intensity includes scattered radiation from the exciting laser at these wavelengths.

will diminish. In presence of laser irradiation for $2p_6 \to 1s_5$, $2p_6 \to 1s_4$, $2p_8 \to 1s_5$, $2p_8 \to 1s_4$ and $2p_9 \to 1s_5$ transitions decrease in intensity is observed, where a reduction in the population of $2p_j$ manifolds due to above process is large as compared to collisional de-excitation from $2p_2$ to these states. This decrease in intensity under laser excitation is predominant in the case of $2p_8 \to 1s_4$ and $2p_9 \to 1s_5$ transitions as shown in figure 2. However such decrease in population density in $2p_j$ levels ($j = 1, 3, 4, 5$) will be compensated by non-radiative transitions from $2p_2$ state. This fact is in support of the observations that the emission intensity for transitions from $2p_j$ levels ($j = 1, 3, 4, 5$) do not have any decrease under laser excitation. An exception is observed in the case of $2p_7 \to 1s_5$ transition where there is an enhancement in intensity at higher current.

Laser excitation of $1s_5 \to 2p_4$ transition (594·5 nm)

The emission and the laser-induced fluorescence spectrum of the neon discharge in the wavelength range from 580 nm to 670 nm at a discharge current of 5 mA are shown in figure 4. The emission intensity for both conditions varies linearly with the discharge current for all the observed transitions within this spectral region. The

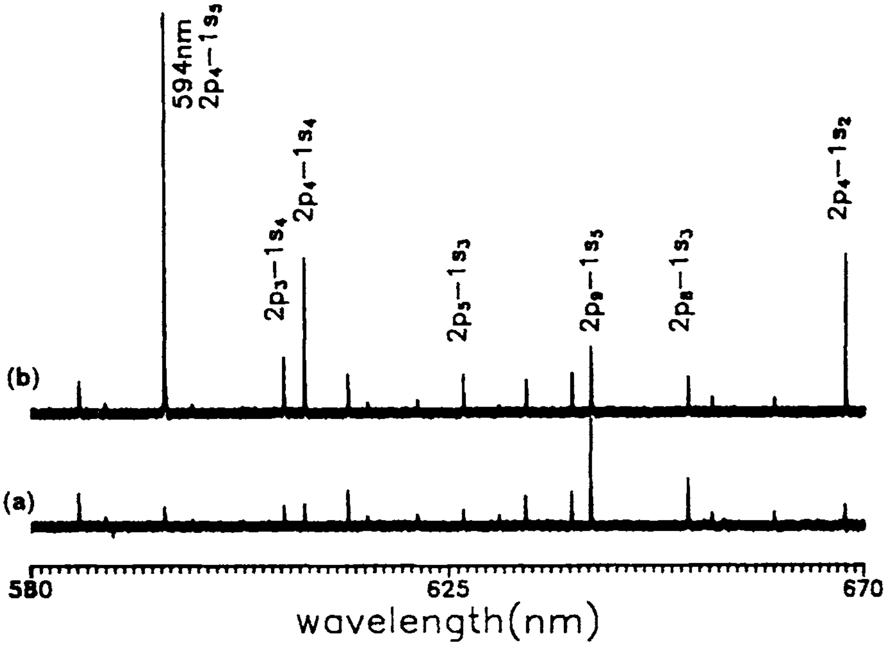

Figure 4. a) Emission spectrum of neon discharge at 5 mA and b) the same under laser irradiation at 594·5 nm.

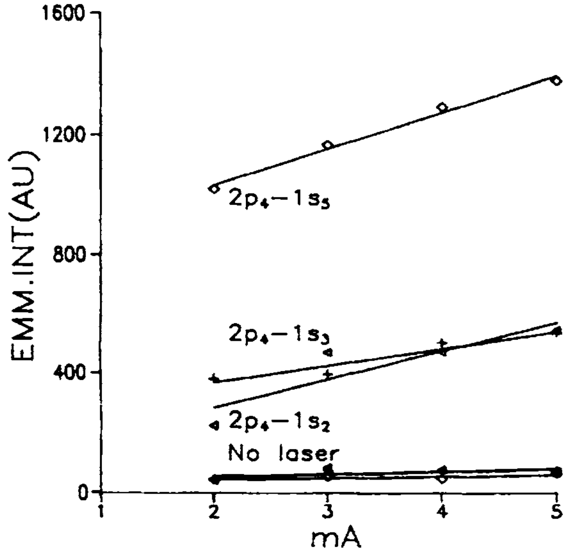

Figure 5. Dependence of the emission intensity for $2p_4 \rightarrow 1s_i$ ($i = 2, 4, 5$) transitions on discharge current.

transitions having considerable change in emission intensities during laser-on and laser-off conditions are shown in the figure 5. Laser excitation at 594·5 nm ($1s_5 \rightarrow 2p_4$) modifies the emission characteristics of transitions from $2p_4 \rightarrow 1s_i$ ($i = 2, 4$ and 5) which is obviously due to a large increase in the population of $2p_4$. For $2p_3 \rightarrow 1s_4$ and $2p_5 \rightarrow 1s_3$ transitions also the same behavior is observed while for $2p_8 \rightarrow 1s_3$, and

$2p_9 \rightarrow 1s_5$ the emission intensity during laser excitation is less than that of the unperturbed discharge. For all other transitions intensities under both conditions are more or less the same. In this case also as it is clear from table 1 that for states close to $2p_4$ the population density increases.

c) *Optogalvanic signal*

The observed OG signal for these transitions as a function of the discharge current is shown in figure 6. It is to be noted that the OG signal was measured at the line centre of these transitions by tuning the laser with an accuracy of 0·01 cm^{-1}. It is well-known that the nature of OG signal is determined mainly by the metastable concentrations. In general, the impedance of a neon discharge increases with resonant excitation of transitions starting from the $1s_3$ and $1s_5$ levels due to the depletion in these metastables and consequently in electron density and the impedance decreases in the case of transitions from $1s_2$ and $1s_4$ states which may decay radiatively to the ground state [4, 6]. Excitation at 588·2 nm and 594·5 nm corresponds to $1s_5 \rightarrow 2p_2$ and $1s_5 \rightarrow 2p_4$ transition respectively where the lower state is metastable and hence the OG signals of these transitions will be an increase in the discharge impedance or will be an equivalent decrease in current in the circuit.

These observations indicate that the major factor which influences the generation of the OG signal is the modifications in the ionization processes as a result of the perturbations in the populations of states. The laser excitation at the above wavelengths result in an increase in the population of $2p_2$ (or $2p_4$), $2s_2$ and $1s_4$ while it decreases in the case of $1s_5$ density. Thus the contributions of $1s_2$, $1s_4$ and the upper state ($2p_2$ or $2p_4$) states, which have strong radiative coupling to the lower states, are negligible whereas the role of $1s_3$ and $1s_5$ metastables are prominent in generating the OG effect. As shown in figure 6, the current dependence of OG signal corresponding to 588·2 nm and 594·5 nm differ widely. This indicates that apart from the metastable nature of the lower level, there exists other processes which will affect magnitude of the OG signal. One such process is due to the change in ionization rate as a result of collisional mixing between $2p_j$ states followed by laser irradiation which is different for these two transitions.

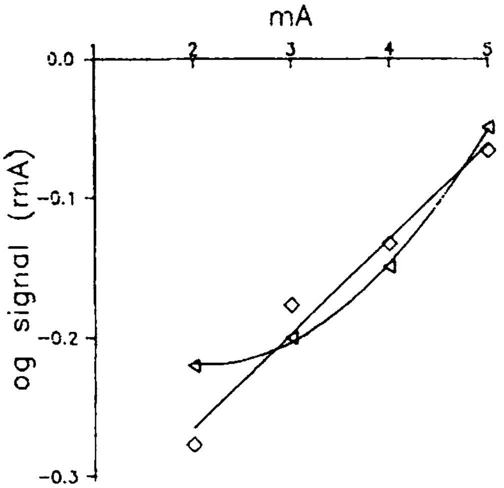

Figure 6. OG signal for $1s_5 \rightarrow 2p_2$ (□) and $1s_5 \rightarrow 2p_4$ (Δ) transition.

4. Conclusions

Modifications in the emission characteristics of a neon hollow cathode discharge due to the perturbation caused by resonant absorption of $1s_5 \rightarrow 2p_2$ and $1s_5 \rightarrow 2p_4$ transition are investigated. In both cases the emission properties are found to be altered significantly due to the changes in the population of $2p$ state by radiative and non-radiative processes. The population of metastable lower state ($1s_5$) and the nature of excitation/de-excitation path ways play an important role in generating the OG signal.

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